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# Magnetic properties of GdCo-Al<sub>2</sub>O<sub>3</sub> composite films

A N Gorkovenko<sup>1,3</sup>, V N Lepalovskij<sup>1</sup> and V O Vas'kovskiy<sup>1,2</sup>

<sup>1</sup>Ural Federal University, Department of Magnetism and Magnetic Nanomaterials,  
620002 Ekaterinburg Mira street 19, Russia

<sup>2</sup>Institute of Metal Physics UB RAS, 620108 Ekaterinburg, S. Kovalevskoy street 18,  
Russia

<sup>3</sup>a.n.gorkovenko@urfu.ru

**Abstract.** The purpose of this work was to synthesize and study magnetic properties of composite films consisting of Gd-Co ferrimagnetic system and Al<sub>2</sub>O<sub>3</sub> dielectric matrix. GdCo-Al<sub>2</sub>O<sub>3</sub> films of different composition were synthesized by magnetron co-sputtering of Gd, Co, and Al<sub>2</sub>O<sub>3</sub> targets in an Ar atmosphere. Investigation of magnetic properties was carried out using a vibrating sample magnetometer in the temperature range from 10 to 300 K. The obtained results showed that in the composite system with a volume of dielectric material exceeding 50% a superparamagnetic state is realized, indicating granular microstructure. In this case, the granules show signs of ferrimagnetic ordering.

## 1. Introduction

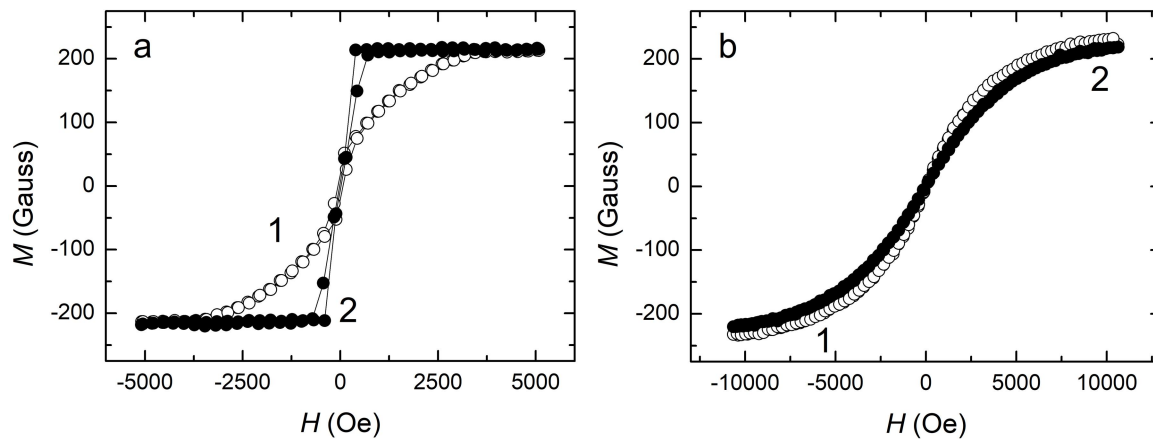
Recently magnetic granular media became a topical direction of research in the physics of magnetic materials. This media represents a variety of composites based on nanometer-sized magnetic particles (granules) imbedded in a non-magnetic matrix. The majority of works in this field is devoted to films in which ferromagnetic metals and iron group alloys act as a metal phase [1, 2]. However, films with granules containing rare-earth elements are potentially richer in terms of the possibilities of tailoring magnetic properties. This work is devoted to the study of the possibilities of obtaining and studying the magnetic properties of granular films based on the Gd-Co ferrimagnetic system and Al<sub>2</sub>O<sub>3</sub> dielectric matrix.

## 2. Experimental

The samples were produced by the magnetron co-sputtering of Gd, Co, and Al<sub>2</sub>O<sub>3</sub> targets on an Orion-8 device in the Ar atmosphere. Due to the low rate of sputtering of Al<sub>2</sub>O<sub>3</sub> compared to metals, in order to obtain a given composition, two Al<sub>2</sub>O<sub>3</sub> targets were sputtered simultaneously. The composite (Gd<sub>y</sub>Co<sub>100-y</sub>)(Al<sub>2</sub>O<sub>3</sub>)<sub>100-x</sub> films with 100 nm of thickness were deposited onto Corning glass substrates and protected by Ta(5 nm) seed and cap layers. The deposition of all layers took place in the presence of a uniform technological magnetic field of 250 Oe applied parallel to the plane of the substrate. The composition of the obtained films was varied by varying the power applied to the corresponding targets. Gd content was varied from 14 to 49% and verified using a Nanohunter X-ray fluorescence spectrometer. The relative volume of the metal phase in the composites was varied as well (from 0.5 to 0.9). It was set according to the predetermined deposition rates of the metal and dielectric components. In addition, to evaluate the effect of composition on the magnetism of a metallic binary system, a series of Gd<sub>y</sub>Co<sub>100-y</sub> films without a dielectric component was obtained. These samples also had a two-sided protective coating, but had a smaller thickness (40 nm).



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**Figure 1.** Hysteresis loops of  $(\text{Gd}_{18}\text{Co}_{82})_{90}(\text{Al}_2\text{O}_3)_{10}$  (a) and  $(\text{Gd}_{29}\text{Co}_{71})_{50}(\text{Al}_2\text{O}_3)_{50}$  (b) films measured parallel (curves 1) and perpendicular (curves 2) the plane of the films.

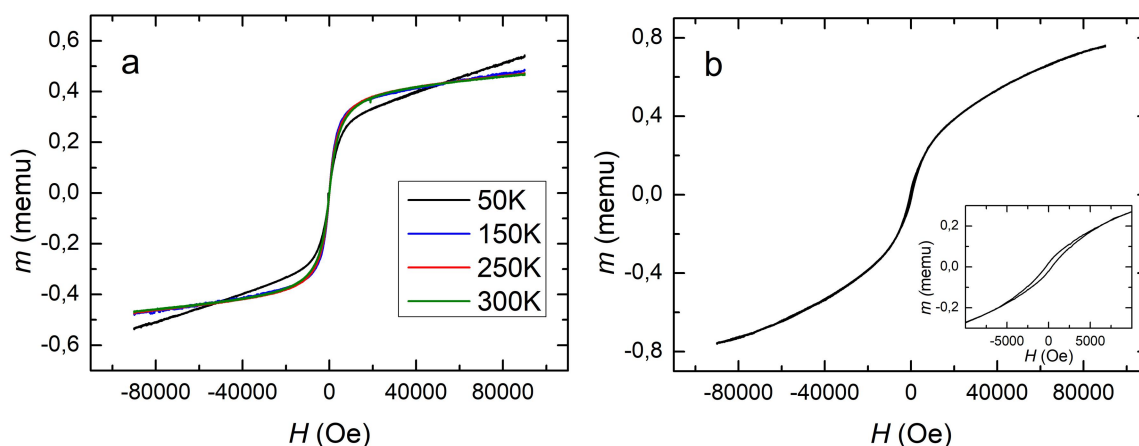
Investigation of magnetic properties of composite films was carried out basing on the analysis of hysteresis loops, which were measured both perpendicular (N type loops) and parallel to the samples plane. In the last case along (type A) and perpendicular (type P) the axis of the technological field applied during the deposition. At room temperature, hysteresis loops were measured using a vibrating sample magnetometer in the field range up to 18 kOe. Temperature studies were carried out using a vibrating set-up of the PPMS DynaCool device, in the temperature range from 10 to 300 K. With the use of this equipment, measurements were also carried out according to the ZFC/FC method. This technique is quite common for the investigation of the magnetic properties of granular media [3,4]. It consists of obtaining the temperature dependences of the magnetizations measured when the sample is heated in a weak magnetic field from two different initial states. The first state is achieved by cooling the sample without an external magnetic field (Zero Field Cooling), the second one - by cooling in a strong magnetic field (Field Cooling). In our case, the field strength during cooling and measurements of the magnetic moment was 70 and 1 kOe, respectively.

### 3. Results and discussion

Figure 1 shows examples of N type (curves 1) and A (curves 2) hysteresis loops for  $(\text{Gd}_{18}\text{Co}_{82})_{90}(\text{Al}_2\text{O}_3)_{10}$  (a) and  $(\text{Gd}_{29}\text{Co}_{71})_{50}(\text{Al}_2\text{O}_3)_{50}$  (b) films. It can be seen that at a relatively low concentration of  $\text{Al}_2\text{O}_3$ , the composite is in magnetically ordered state. Moreover, a relatively small magnitude of spontaneous magnetization indicates the presence of ferrimagnetism in the binary Gd–Co system. In addition, the film shows a so-called perpendicular magnetic anisotropy, i.e. axis of the uniaxial anisotropy (easy magnetization axis or EA) is perpendicular of the film plane. Note that in the homogeneous (purely metallic)  $\text{Gd}_{18}\text{Co}_{82}$  film, the perpendicular anisotropy was not observed. It also had a uniaxial magnetic anisotropy, but with an EA being parallel to the axis of application of the technological field. It is possible that  $\text{Al}_2\text{O}_3$  contributes to the formation of a specific (columnar) microstructure, which is considered to be the main source of perpendicular anisotropy in amorphous Gd–Co films [5].

With the increase in the concentration of the metallic phase up to 50%, all types of loops become nonhysteretic and almost identical in shape, which is clear evidence of the superparamagnetic state of the composite [6,7]. This allows us to conclude that a highly dispersed magnetic system was formed in the film, in other words, a granular structure.

The results of the temperature study of the hysteresis properties of the  $(\text{Gd}_{29}\text{Co}_{71})_{50}(\text{Al}_2\text{O}_3)_{50}$  film are shown in figure 2. In a wide temperature range  $T$ , down to 50 K, the loops are nonhysteretic (figure 2 (a)). A distinct hysteresis is observed only at  $T = 10$  K, which is shown for greater clarity in the inset of figure 2 (b). The appearance of hysteresis at low temperatures is a characteristic of a granular



**Figure 2.** Hysteresis loops of the  $(\text{Gd}_{29}\text{Co}_{71})_{50}(\text{Al}_2\text{O}_3)_{50}$  sample, measured at temperatures of 50, 150, 250 and 300 K (a); 10 K (b). In the insert is a fragment of a hysteresis loop, measured at a temperature of 10 K.

magnetic systems [8] and usually is due to an increase of magnetic anisotropy and the transition of granules to a ferromagnetic state.

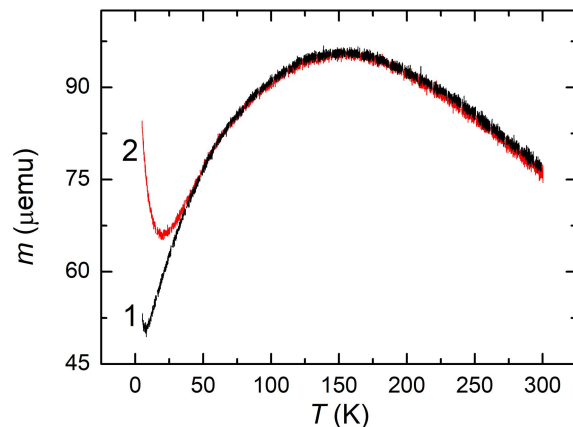
It is also worth noting the almost complete coincidence of hysteresis loops in the temperature range of 100–300 K. This fact requires special study, since in homogeneous metallic films of similar composition, there is a significant increase in spontaneous magnetization with temperature decreasing. It seemed that this would have led to an increase in the initial magnetic susceptibility with temperature decreasing. But it is not observed in the experiment. Moreover, at a temperature of less than 100 K, the loops become flatter, i.e., the susceptibility decreases. But then the maximum magnitude of the magnetic moment increases. It can be assumed that all these features of magnetization, one way or another, reflect a certain balance between temperature changes of spontaneous magnetization and magnetic anisotropy of granules.

Additional information on this subject can be found in the ZFC/FC data. Figure 3 shows the temperature dependences of the magnetic moment  $m$  of the sample  $(\text{Gd}_{29}\text{Co}_{71})_{50}(\text{Al}_2\text{O}_3)_{50}$ , measured in a relatively weak magnetic field (1000 Oe) when heated up from two different magnetic states. They are quite specific. First of all, it refers to the curve FC. It has a pronounced non-monotonic character, whereas for ferromagnetic granular films the monotonously decreasing dependence  $m(T)$  is typical. It suggests a possible magnetic compensation in the region of low temperatures. In amorphous Gd–Co films, compensation near  $T \sim 10$  K can actually be observed [9], but for a significantly lower Gd content. It cannot be excluded that in the process of obtaining composite films, a part of Gd does not bind with Co in a single cluster, but forms a separate metallic or oxide phase. The same applies to Co. Obviously, phase analysis is not possible using the Nanohunter elemental analyzer used.

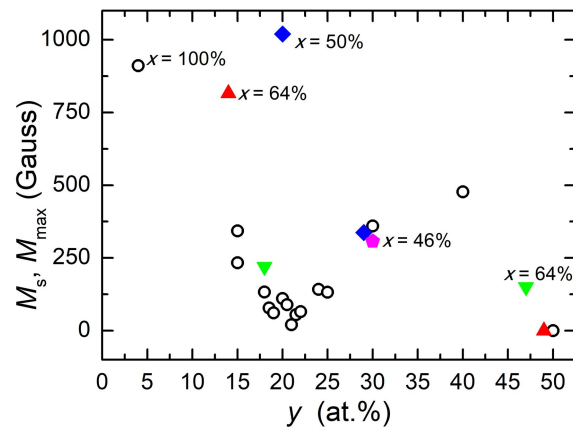
Nevertheless, the ZFC/FC dependences demonstrate an obvious sign of the transition between the ferromagnetic and superparamagnetic states. From figure 3 it can be seen that they coincide in the temperature range of 40–300 K, and diverge at  $T < 40$  K, thereby showing the appearance of magnetic anisotropy. Thus, it can be assumed that at  $T < 40$  K the sample is in the ferromagnetic state, and at  $T > 40$  K it is in the superparamagnetic state.

Returning to the question of the magnetic state of Gd, it should be noted that the compounds  $\text{Gd}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3$  have similar formation enthalpies. This means that oxygen in the process of film formation is equally beneficial to interact with both Gd and Al. However, this is important only under the condition that in the process of sputtering a dielectric target, Ar ions not only knock out  $\text{Al}_2\text{O}_3$  molecules, but also split them. The possibility of the latter scenario is indicated by report [10].

For more information on the magnetism of the composites under study, the concentration dependences of the saturation magnetization  $M_s$  for  $\text{Gd}_y\text{Co}_{100-y}$  films and the maximum magnetization  $M_{\text{max}}$  in a 10 kOe field for  $(\text{Gd}_y\text{Co}_{100-y})_x(\text{Al}_2\text{O}_3)_{100-x}$  samples were compared. Note that in the last case,



**Figure 3.** Temperature dependences of the magnetic moment of the  $(\text{Gd}_{29}\text{Co}_{71})_{50}(\text{Al}_2\text{O}_3)_{50}$  film, obtained in the ZFC (curve 1) and FC (curve 2) modes. The applied magnetic field was 1000 Oe.



**Figure 4.** Dependences of the saturation magnetization of the  $\text{Gd}_y\text{Co}_{100-y}$  films and the metal phase in  $(\text{Gd}_y\text{Co}_{100-y})_x(\text{Al}_2\text{O}_3)_{100-x}$  films as a function of the Gd content.

$M_{\text{max}}$  either coincided with the saturation magnetization (for films with a relatively low dielectric content), or was estimated to be close to it (for samples in a superparamagnetic state).

In figure 4, the  $M_s$  values for  $\text{Gd}_y\text{Co}_{100-y}$  films are shown with hollow points. The filled points correspond to the normalized magnetization  $M_{\text{max}}$  values of the  $(\text{Gd}_y\text{Co}_{100-y})_x(\text{Al}_2\text{O}_3)_{100-x}$  films. Normalized values of magnetization were obtained by dividing the measured magnetic moment by the nominal volume of the metallic phase. It can be seen that the dependences  $M_s(y)$  and  $M_{\text{max}}(y)$  correlate well enough and in both cases are non-monotonic, indicating the presence of magnetic compensation at  $y \sim 20$  at.% Gd. This indirectly indicates the absence of phase separation in the metallic component of the composite.

#### 4. Conclusion

In this work we demonstrated the possibility of producing granular  $(\text{Gd}_y\text{Co}_{100-y})_x(\text{Al}_2\text{O}_3)_{100-x}$  magnetic films by ion cosputtering of the Gd, Co,  $\text{Al}_2\text{O}_3$  targets. It is shown that at  $y \geq 50$  a superparamagnetic state is realized in composite films. The presence of ferrimagnetic state of granular structure was shown.

#### Acknowledgment

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